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Rule of Thumb for Estimating Groundwater Activation from Residual Dose Rates

J. Donald Cossairt
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Author: J. Donald Cossairt Date: 6/5/98
J. Donald Cossairt

Reviewed: Date: 6/5/98
K. Vaziri

Approved:: Date: 6/9/98
R. Walton
Associate Head, Environmental Protection

Approved: Date: 6/9/98
Bill Griffing
Head, ES&H Section

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There has long been an informal "rule of thumb" at Fermilab that states that if the maximum residual absorbed dose rate external to some beam element is less than 100 mrads/hour, there is no significant risk of contamination of groundwater resources with radionuclides above the permissible limits. Indeed, this Rule of Thumb may date back to the late Miguel Awschalom. This criterion, as it is generally quoted, does not specify any details as to irradiation time, decay time, or distance from the activated beamline component. The purpose of this note is to investigate this Rule of Thumb. This exercise was done primarily to investigate the validity of this undocumented "rule" which had formerly existed as "lore" in the minds of some of the more senior physicists at the Laboratory. For specific situations where the usage of this criterion leads one to even remotely suspect the presence of significant soil activation, a separate and more detailed investigation should be performed. It is intended to be used only as a preliminary screening tool.

The approach taken is to use the residual dose rate to derive an estimate of the flux density of energetic particles presented to the enclosure wall while the beamline is operating. Using that flux density, then, one can derive the concentration of radionuclides present in the media external to the wall by employing elements of the so-called Concentration Model (Wehmann, et al, 1993, Malensek, et al, 1993, and Cossairt, 1994).

Following P. Gollon (1976), one has

$$\frac{dD}{dt} = G_c \frac{dS_{Fe}}{dt} \omega(t_i, t_c) , \qquad (1)$$

where dD/dt is the absorbed dose rate (mrad hr^{-1}) at some distance from the surface of a component. dS_{Fe}/dt (stars cm^{-3} s^{-1}) is the rate of star production per unit volume per unit time at the outer surface of a typical beamline component made of iron as might be calculated by a standard high energy Monte-Carlo code such as CASIM (Van Ginneken and Awschalom, 1975) or MARS (Mokhov, 1998). The factor $\omega(t_i, t_c)$ is a function of the irradiation time, t_i, and the cooldown time, t_c , respectively, that takes into account the decay of the aggregate of the radionuclides produced in the beamline component. It is derived from the so-called "danger parameter" of M. Barbier (1969) and conventionally has the units of mrad hr^{-1} per star cm^{-3} s^{-1}. The factor G_c is the fractional solid angle subtended by the beamline component at the location of the "observer" and obviously has a maximum value of 0.5 for an observer next to an infinite wall. Gollon gives, for iron components;

$$\omega(\infty,0) = 9\times10^{-3} \ \text{(mrad hr}^{-1}\text{ per star cm}^{-3}\text{ s}^{-1}\text{) and}$$
$$\omega(30d,1d) = 2.5\times10^{-3} \ \text{(mrad hr}^{-1}\text{ per star cm}^{-3}\text{ s}^{-1}\text{).} \qquad (2)$$

While Gollon gives the value of $\omega(t_i, t_c)$ for only two pairs of irradiation and decay times, it clearly would be useful to have others. This has been done by returning to the early references of the Oak Ridge group; namely, Armstrong and Alsmiller (1969) and Gabriel and Santoro (1973). Gollon derived his values of $\omega(t_i, t_c)$ from this general work of the Oak Ridge group. Figures 1 - 3 present the results of obtaining values of $\omega(t_i, t_c)$ by scaling the results of the Oak Ridge Group for irradiations, t_i, of one month, one year, and an "infinitely" long time to Gollon's value for $\omega(\infty,0)$. It is not at all surprising that Gollon's value for ω(30d,1d) (Fig. 1) is in good agreement with the result scaled from the other two publications since these works form the basis of Gollon's. In the rest of the work presented here, the values of $\omega(t_i, t_c)$ derived from Armstrong and Alsmiller will be used since they cover a larger domain of t_c .

One may now solve eqn (1) for dS_{Fe}/dt if one has some idea of the values of t_i and t_c that are associated with the measurement of the absorbed dose rate dD/dt ,

$$\frac{dS_{Fe}}{dt} = \frac{1}{G_c\omega(t_i,t_c)}\frac{dD}{dt}. \qquad (3)$$

Based on this value of dS_{Fe}/dt at the outer surface of the beam line component, one needs to get an estimate of the average star density production rate at the surface of the wall during the irradiation. One can do this by observing that the flux density of particles is conserved when crossing material boundaries. The flux density of particles considered here, ϕ_H, is that of the particles having momenta above the lower momentum cutoff of 300 MeV/c (47 MeV nucleon kinetic energy) conventionally used in the code CASIM. By coincidence, the approximate production thresholds of radionuclides of concern in groundwater roughly correspond to the CASIM lower momentum cutoff. ϕ_H does not include, for example, low energy neutrons that might impinge upon the enclosure walls but would produce no radioactivity of significance beyond the walls. The conservation of ϕ_H can be used to provide a connection between the production of stars in the beamline component and the consequent production of radioactivity, in the enclosure walls and beyond.

At any point in a material medium ϕ_H is related to the star density production, dS/dt, at that same point through the nonelastic interaction length for that material, λ, by means of* ;

$$\phi_H = \lambda \frac{dS}{dt}. \tag{4}$$

Thus, applying this conservation of ϕ_H, the star density production at the wall, dS_{wall}/dt will be related to that near the surface of the iron beamline component, dS_{Fe}/dt, by means of:

$$\lambda_{wall} \frac{dS_{wall}}{dt} = G_w \lambda_{Fe} \frac{dS_{Fe}}{dt}, \tag{5}$$

where the values of interaction length for various materials, in units of centimeters, can be obtained from the tabulations of the Particle Data Group (1996) using the associated values of material densities. The factor G_w contains any reduction of flux density between the beamline element and the wall and has a maximum value of unity. It is essentially another factor dependent upon solid angle subtended by the beamline component at the wall. G_c and G_w have the same dependence upon the distance from the activated component, which for point-like components is inverse square. Customarily, one takes the concrete of the wall and the media outside it (e.g., soil or rock) to be materials of equivalent composition with slightly different densities. For concrete walls, $\lambda_{Fe}/\lambda_{wall} \approx 0.40$. Substituting, one gets,

$$\frac{dS_{wall}}{dt} = \frac{\lambda_{Fe}}{\lambda_{wall}} \frac{G_w}{G_c \omega(t_i, t_c)} \frac{dD}{dt} \text{ (stars cm}^{-3}\text{ s}^{-1}\text{)}. \tag{6}$$

In general, walls are approximately 30 centimeters (1 ft) thick. To good approximation, this amounts to a factor of two attenuation before the flux of particles produced by the hadronic cascade in the beamline component reaches "unprotected" media, typically soil. The difference between the density of standard Fermilab soil (2.25 g cm^{-3}) and concrete (2.4 g cm^{-3}) is only 6.6 per cent and will be ignored here. Proceeding, the maximum star production rate in the environmental media (soil or rock),dS_{media}/dt, is given by

$$\frac{dS_{media}}{dt} = 0.5 \frac{\lambda_{Fe}}{\lambda_{wall}} \frac{G_w}{G_c \omega(t_i, t_c)} \frac{dD}{dt} \text{ (stars cm}^{-3}\text{ s}^{-1}\text{)}. \tag{7}$$

If one uses "contact" survey readings on the side of the beamline component, G_c can be set equal to 0.5. Thus, the values $G_c = 0.5$ and $G_w = 1$ are chosen. The sensitivity of the results to this choice is not large since a smaller value of G_c would be associated with a correspondingly

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* Care with units of measure must be taken here since λ is commonly expressed in units of g cm^{-2}. If dS/dt is in units of stars cm^{-3}s^{-1}, λ must be in cm in order to obtain ϕ_H in cm^{-2}.

smaller value of G_w. Thus, taking the survey of absorbed dose residual activity at the enclosure wall assures $G_w/G_c \equiv 2$.

dS_{media}/dt, can now be connected to the concentration of radionuclides that might be produced in the media. According to the Concentration Model, the initial concentration of the i^{th} radionuclide, $C_{0,i}$, in the water in the media in the immediate vicinity of the wall after an irradiation time, t, is given by:

$$C_{0,i} = \frac{N_p A S_{\max} K_i L_i}{1.17 \times 10^6 \rho_{media} w_i}[1 - \exp(-t/\tau_i)], \quad \text{(pCi/ml)} \qquad (8)$$

where N_p is the number of protons per year, A is a constant which averages the maximum star density per incident proton, S_{max}, (stars cm^{-3} incident proton^{-1}) over some representative volume of the region near the interaction point (see below), K_i, is the number of atoms of the i^{th} radionuclide produced per star, L_i is the leachability of the i^{th} radionuclide, τ_i is the meanlife (years) of the i^{th} radionuclide, ρ_{media} is the density (g cm^{-3}) of the media, and w_i, is the weight of water needed to leach out the i^{th} radionuclide to the level specified by L_i. The constant in the denominator collects the unit conversions. The factor in square brackets obviously describes the buildup to saturation during irradiations. For irradiation times long compared to τ_i, the term in square brackets becomes unity, and the maximum, or so-called "saturated", value of $C_{0,i}$ is the result. The formula implicitly does not allow for any migration of radionuclides from the vicinity of the source during the irradiation.

The product $N_p S_{max}$ can be determined as follows,

$$N_p S_{\max} = 3.156 \times 10^7 \frac{dS_{media}}{dt}, \qquad (9)$$

where the constant is the number of seconds in a year and dS_{media}/dt is in units of stars cm^{-3}s^{-1}. Thus, combining constants

$$C_{0,i} = \frac{26.97 A K_i L_i}{\rho_{media} w_i} \frac{dS_{media}}{dt}[1 - \exp(-t/\tau_i)] \quad \text{(pCi/ml)}. \qquad (10)$$

When the Concentration Model was first developed, a value of A = 0.019 was found to be appropriate for large beam absorbers typical of the Fixed Target experimental areas at Fermilab. More recently, it has become better appreciated that this value of A is not always valid. For example, for a situation in which the star density is nearly independent of the longitudinal distance from the target, a value A = 0.19 is a better choice (Freeman, 1996). The smaller value is more appropriate for sources that are more "point-like". Thus, taking A = 0.1 appears to be a conservative estimate for most situations. A smaller value of A might be appropriate for many situations as determined by the judgment of the user of this method or, better yet, as the result of a specific Monte-Carlo calculation. As demonstrated by Borak, et al (1972), the only two

radionuclides of concern with respect to protection of groundwater resources are ^{3}H (τ = 17.78 years) and ^{22}Na, (τ = 3.754 years).

It is useful to calculate values of $C_{o,i}$ from a variety of combinations of t_i and t_c and for our referenced residual absorbed dose rate of 100 mrad/hr. The calculation presented here used the "standard" parameters of the Concentration Model (see Cossairt, 1994). Table 1 lists the parameters chosen for this calculation and Table 2 gives the results.

Table 1 Parameters chosen for calculation of $C_{o,i}$ in units as discussed in the text.

Parameter	Value
G_c	0.50
G_w	1.00
$\lambda_{Fe}/\lambda_{wall}$	0.40
$\tau(^{3}H)$	17.7783
$\tau(^{22}Na)$	3.7539
A	0.1000
dD/dt	100
ρ	2.25
$K_{tritium}$	0.075
$L_{tritium}$	0.9
$w_{tritium}$	0.27
K_{sodium}	0.02
L_{sodium}	0.135
w_{sodium}	0.52

Table 2 Calculated values of $C_{o,i}$ for various combinations of t_i and t_c using the parameters of Table 1.

t_i (years)	t_c	$\omega(t_i, t_c)$ (mrad h^{-1} per stars $cm^{-3}s^{-1}$)	$C_{0,3}$ (^{3}H) (pCi/ml)	$C_{0,22}$(^{22}Na) (pCi/ml)
1	1 day	4.98×10^{-3}	131.7	11.7
1	1 week	3.70×10^{-3}	177.2	15.7
1	1 month	2.59×10^{-3}	253.2	22.5
5	1 day	5.84×10^{-3}	503.2	31.4
5	1 week	4.70×10^{-3}	625.3	39.0
5	1 month	3.36×10^{-3}	874.6	54.5
10	1 day	6.70×10^{-3}	769.7	34.6
10	1 week	5.40×10^{-3}	955.0	42.9
10	1 month	4.12×10^{-3}	1251.8	56.2
∞	1 day	6.70×10^{-3}	1789.2	37.2
∞	1 week	5.40×10^{-3}	2219.9	46.1
∞	1 month	4.12×10^{-3}	2909.6	60.4

After calculating this initial concentration, one must make an assumption concerning the migration to the aquifer. Typically, Fermilab beam enclosures are located in the glacial till no more than about 5 meters below grade level. Such enclosures are, then typically several meters above the aquifer of concern. Following the Concentration Model, the concentrations of the two radionuclides of concern as a function of distance, d (meters), between the source and the aquifer are given by reduction factors that have been fitted by exponentials which implicitly include both dispersion and radioactive decay;

$$C_3(d) = C_{0,3}e^{-0.3d} \quad (^3\mathrm{H}) \tag{11}$$

$$C_{22}(d) = C_{0,22}e^{-0.92d} \quad (^{22}\mathrm{Na}). \tag{12}$$

The results for the sets of (t_i, t_c) considered above are plotted in Figures 4 and 5 as a function of depth, d, using these simple exponential fits. These figures are intended to be used as nomographs to obtain a conservative estimate of maximal concentrations in water that might result. Other coefficients of d in the functions above may be used to calculate these concentrations in circumstances where it is believed that they may provide a better representation of the downward migration as discussed by Malensek, et al (1993) and Cossairt (1994).

The calculated results for $C_{0,3}$ and $C_{0,22}$ for long irradiations are somewhat larger than the standards of 2000 pCi/ml and 10 pCi/ml applied to surface water discharges derived from DOE Order 5400.5 (DOE, 1990) under some irradiation conditions. In practice, over the years of operations of the Fermilab accelerators, there have been numerous instances where beamline components have reached 100 mrad/hour residual absorbed dose rate. However, no radionuclide concentrations approaching the levels calculated above have been observed in discharges from the underdrains, a result probably due to a large dilution by rainwater in the vicinity of the various beam enclosures. Furthermore, these "hot spots" are highly localized and often do not persist for lengthy periods of time (years).

The results at the aquifer must be compared with the corresponding concentrations for groundwater resources specified by State regulation and DOE Orders by means of a weighted sum. These values are taken to be 20 pCi/ml for ^{3}H and 0.4 pCi/ml for ^{22}Na (Cossairt, 1994) and compliance is to be demonstrated by assuring that the sum of the concentrations each divided by the regulatory standard is less than unity. For ^{22}Na, concentrations below 0.4 pCi/ml are achieved for all irradiation conditions at a depth, d, of about 6 meters or more. For ^{3}H, values below 20 pCi/ml are achieved at depths greater than 17 meters for all irradiation conditions. For values of d from 10 to 15 meters, typical of Fermilab target stations, ^{3}H concentrations exceeding 20 pCi/ml are achieved for irradiations longer than 5 years.

Conclusion

Using the parameters assumed in this paper, the rule of thumb is verified for irradiations of some, but not all durations. For example, if the irradiations are less than about 5 years and the source is more than 13 meters above the aquifer the groundwater standards are met. For longer

irradiations, the aquifer must be deeper than 14 meters below the source (10 years irradiation) or 17 meters below the source ("infinite" irradiation). The upper aquifers at Fermilab are typically closer to the surface than this value. Thus, this rule of thumb should be used cautiously, and with a full appreciation for the effects of the uncertainties of the chosen parameters. It is preferred by far to do:

1) A complete Monte Carlo calculation for suspect locations to determine the product AS_{max}; and

2) A complete calculation of the propagation of radionuclides to the aquifer by the use of, e.g., PATCH3D (Sudicky, 1988).

In both cases, the best available local physical and hydrogeologic data should be used.

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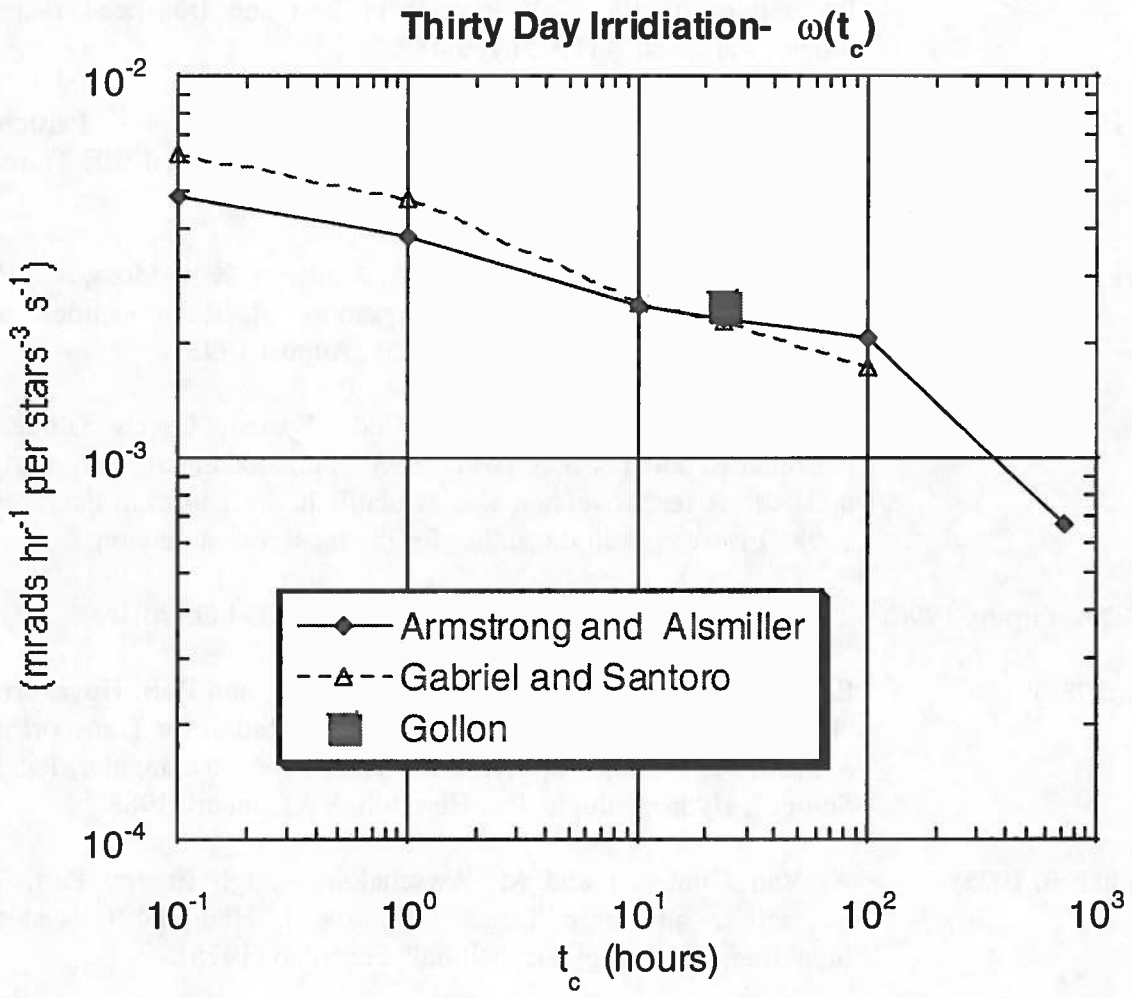
Thirty Day Irridiation- ω(t_c)
10^-2
10^-3
10^-4
(mrads hr^-1 per stars^-3 s^-1)
Armstrong and Alsmiller
Gabriel and Santoro
Gollon
10^-1
10^0
10^1
10^2
10^3
t_c (hours)

Figure 1

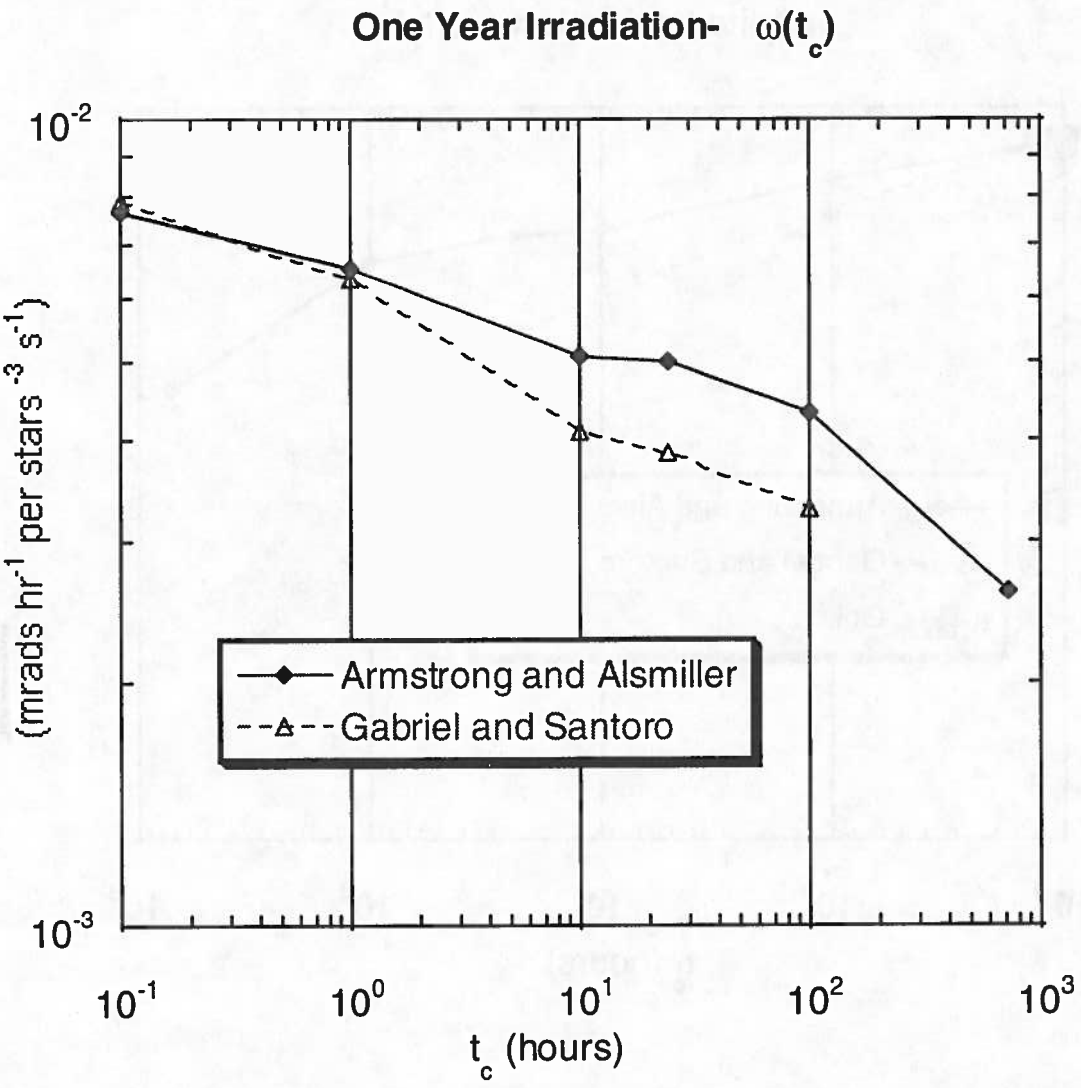
One Year Irradiation- ω(t_c)
10^-2
10^-3
(mrads hr^-1 per stars^-3 s^-1)
Armstrong and Alsmiller
Gabriel and Santoro
10^-1
10^0
10^1
10^2
10^3
t_c (hours)

Figure 2

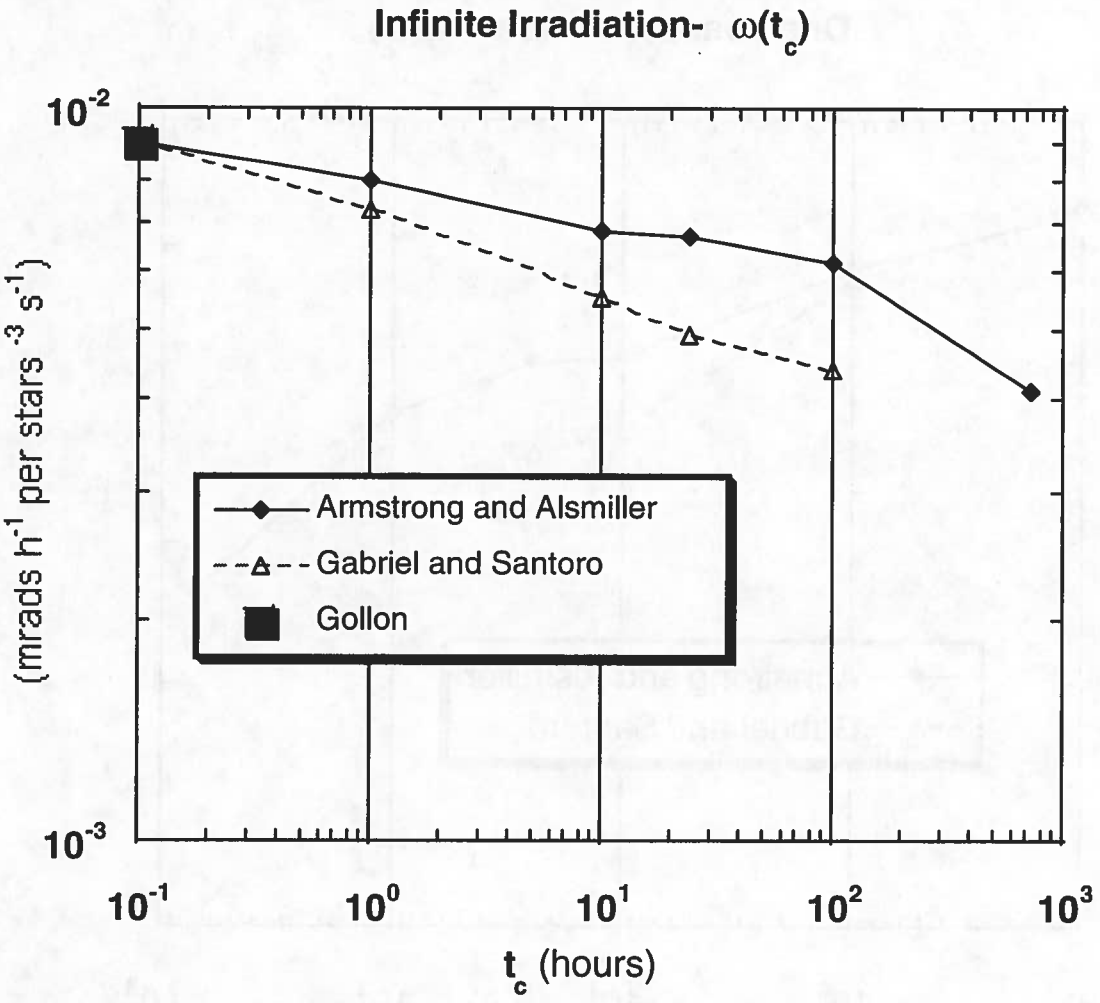
Infinite Irradiation- ω(t_c)
10^-2
10^-3
(mrads h^-1 per stars ^-3 s^-1)
Armstrong and Alsmiller
Gabriel and Santoro
Gollon
10^-1
10^0
10^1
10^2
10^3
t_c (hours)

Figure 3

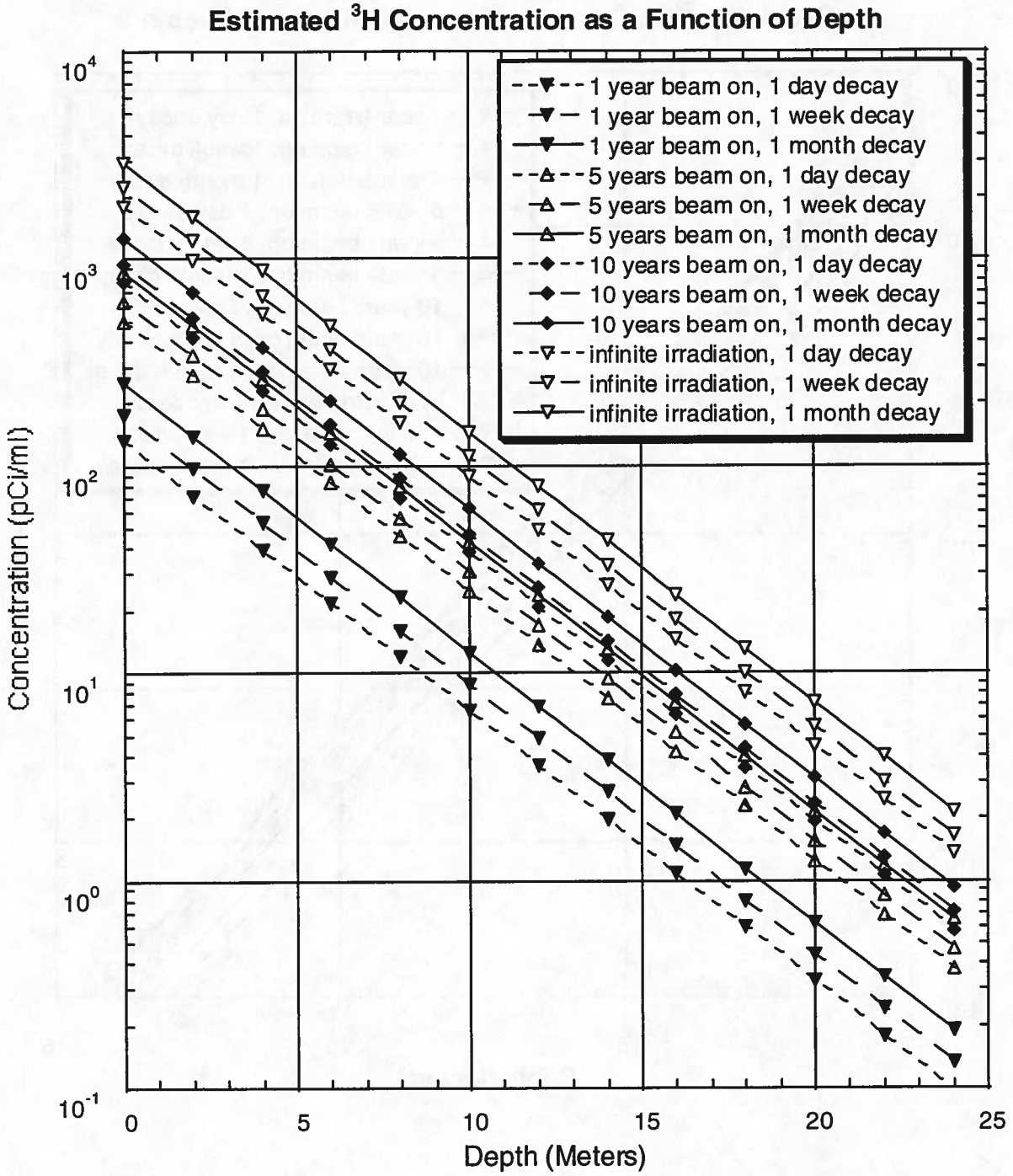
Estimated ^{3}H Concentration as a Function of Depth
1 year beam on, 1 day decay
1 year beam on, 1 week decay
1 year beam on, 1 month decay
5 years beam on, 1 day decay
5 years beam on, 1 week decay
5 years beam on, 1 month decay
10 years beam on, 1 day decay
10 years beam on, 1 week decay
10 years beam on, 1 month decay
infinite irradiation, 1 day decay
infinite irradiation, 1 week decay
infinite irradiation, 1 month decay
Concentration (pCi/ml)
10^{4}
10^{3}
10^{2}
10^{1}
10^{0}
10^{-1}
0
5
10
15
20
25
Depth (Meters)

Figure 4

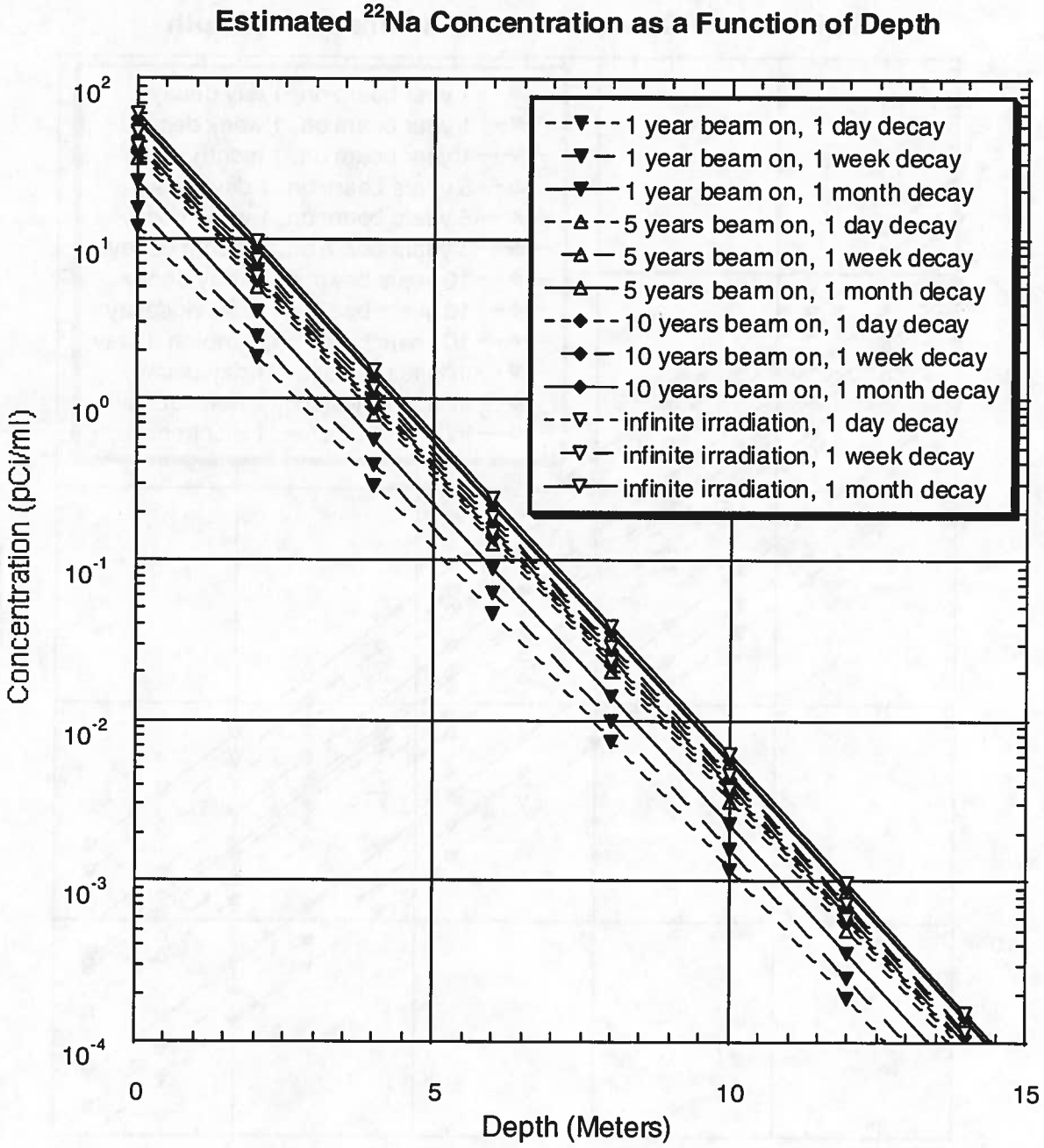
Estimated ^{22}Na Concentration as a Function of Depth
Concentration (pCi/ml)
10^{2}
10^{1}
10^{0}
10^{-1}
10^{-2}
10^{-3}
10^{-4}
0
5
10
15
Depth (Meters)
1 year beam on, 1 day decay
1 year beam on, 1 week decay
1 year beam on, 1 month decay
5 years beam on, 1 day decay
5 years beam on, 1 week decay
5 years beam on, 1 month decay
10 years beam on, 1 day decay
10 years beam on, 1 week decay
10 years beam on, 1 month decay
infinite irradiation, 1 day decay
infinite irradiation, 1 week decay
infinite irradiation, 1 month decay

Figure 5